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Numerical simulation of helical waves arising in self-propagating high-temperature syntheses

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Self-propagating High-temperature Syntheses (SHS)

Self-propagating High-temperature Syntheses (SHS) recently attract a great deal of attention as a synthetic method creating refractory ceramics, intermetallic compounds, composites and so on ([9] for instance). Among over 100 kinds of SHS, a typical example is the synthesis of titanium carbide TiC from titanium Ti and carbon C. In the process, we ignite the top end of cylindrical sample formed by compressing powdery raw materials, say Ti and C, and the temperature there goes up to over 3,000 K by a great quantity of heat emitted along with chemical reactions between raw materials in the sample. Thus a combustion disk appears and the synthesis spreads to the neighborhood warmed by the heat diffusion effect. Once the top end of compact is set fire to, then a reaction front propagates along the compact with almost no energy supply and the synthesis in the compact of 4 cm long completes in a few seconds.

When a combustion wave keeps its profile and propagates at a constant velocity, one can create a very-high-quality uniform product by the SHS. In other words, combustion waves of steady-state mode bring high-quality products. When experimental conditions are changed, however, the combustion wave of steady-state mode may lose its stability and gives place some non-uniform combustion wave, which deteriorates quality. Actually, it is reported that a pulsating combustion wave emerges through the Hopf bifurcation of a combustion wave of steady-state mode ([9]). We also observe a wave that propagates in the form of spiral encircling the cylindrical sample with several reaction spots, which is called a helical combustion wave in the present paper. We note that similar helical waves are reported in autocatalytic reactions and propagation fronts of polymerizations (Pojman et al. [10]).

In the present paper, by using mathematical models for the SHS process, we try to clarify the existing condition of stable helical wave and transition processes of wave patterns from steady-state mode to helical mode via pulsating ones. We have studied numerically the behaviors of propagating waves in cylindrical domains and have obtained the following results:

(a) Set physical parameters so that a pulsating wave exists stably for the one-dimensional problem. Then, the planar pulsating wave is still stable in the cylindrical domain when the radius is small while a helical wave takes the place of the pulsating wave when the radius becomes larger.

(b) When physical parameters are fixed so that a combustion wave of steady-state mode is stable in the one-dimensional problem, the planar wave of steady-state mode is stable also

in the cylindrical domain.

A dimensionless mathematical model for SHS

We begin with a mathematical model for a two-component spatially uniform synthesis



where R_1 and R_2 denote reactants, P a product and m and n reaction orders. The chemical reaction rate $k(T)$ is a function of temperature T governed by the Arrhenius law, and it is expressed as

$$k(T) = k_0\phi(T), \quad \phi(T) = \begin{cases} \exp(-E/RT) & T \geq T_{ig}, \\ 0 & T < T_{ig} \end{cases} \quad (2)$$

by the use of a pre-exponential constant k_0 , an effective activation energy E , the universal gas constant $R = 8.3135 \text{ J/mol}\cdot\text{K}$ and an ignition temperature T_{ig} . This form of $k(T)$ means that no reaction occurs when T is less than T_{ig} and the synthesis starts when T goes up to T_{ig} . Under the assumption $\frac{1}{m}[R_1] = \frac{1}{n}[R_2]$, where $[R_1]$ and $[R_2]$ denote the initial concentration of reactants R_1 and R_2 , respectively, the model system of equations is given by

$$\begin{cases} C_p\rho\dot{T} = Qk_0m^mn^nc^{m+n}\phi(T), \\ \dot{c} = -k_0m^mn^nc^{m+n}\phi(T). \end{cases} \quad t > 0, \quad (3)$$

where the dot means the differentiation by time t , and the unknown variables are the temperature $T = T(t)$ and $c(t) = \frac{1}{m}$ (the concentration of R_1) at time t . Here, C_p denotes the specific heat capacity, ρ the density of compact and Q the reaction heat.

In general, both C_p and Q depend on the temperature, however, we assume them to be constant in the present paper by taking their suitable average. Thus, integrating the both hand side of $C_p\rho\dot{T} = -Q\dot{c}$, we can define the burned temperature T_b by

$$T_b = T_0 + \frac{Q}{C_p\rho}c_0 \quad (4)$$

where $T_0 = T(0)$ and $c_0 = c(0)$ are the initial temperature and concentration, respectively. We note that the solution $(T(t), c(t))$ of (3) tends to $(T_b, 0)$ as $t \rightarrow \infty$ if $T_0 \geq T_{ig}$.

Considering the spatial distribution $T(t, \mathbf{x})$ and $c(t, \mathbf{x})$ of temperature and reactant concentration and introducing the heat diffusion effect, we arrive at the following mathematical model for SHS:

$$\begin{cases} C_p\rho\frac{\partial T}{\partial t} = C_p\rho\alpha\Delta T + Qk_0m^mn^nc^{m+n}\phi(T), \\ \frac{\partial c}{\partial t} = -k_0m^mn^nc^{m+n}\phi(T), \end{cases} \quad \text{in } \Omega, \quad t > 0, \quad (5)$$

where α is a diffusion constant and Ω denotes the cylindrical domain occupied by the sample compact. Introducing dimensionless parameters and variables defined by

$$\begin{cases} K_0 = k_0 m^m n^n c_0^{m+n-1}, \quad \tau = K_0 t, \quad \mathbf{y} = \sqrt{\frac{K_0}{\alpha}} \mathbf{x}, \\ u(\tau, \mathbf{y}) = \frac{T(\frac{\tau}{K_0}, \sqrt{\frac{\alpha}{K_0}} \mathbf{y}) - T_0}{T_b - T_0}, \quad \eta(\tau, \mathbf{y}) = 1 - \frac{1}{c_0} c(\frac{\tau}{K_0}, \sqrt{\frac{\alpha}{K_0}} \mathbf{y}) \end{cases} \quad (6)$$

and rewriting τ , \mathbf{y} and $\sqrt{K_0/\alpha}\Omega = \{\sqrt{K_0/\alpha}\mathbf{x}; \mathbf{x} \in \Omega\}$ respectively as t , \mathbf{x} and Ω , we derive the following non-dimensional system from (5):

$$\begin{cases} \frac{\partial u}{\partial t} = \Delta u + (1 - \eta)^N \Phi(u), \\ \frac{\partial \eta}{\partial t} = (1 - \eta)^N \Phi(u), \end{cases} \quad \text{in } \Omega, \quad t > 0. \quad (7)$$

The new variables u and η are called non-dimensional temperature and a synthesis rate, respectively. In (7), $N = m + n$ and $\Phi(u)$ is expressed as

$$\Phi(u) = \begin{cases} \exp(-\frac{e}{u + u_0}) & u \geq u_{ig}, \\ 0 & u < u_{ig}, \end{cases} \quad (8)$$

by using non-dimensional parameters

$$e = \frac{E}{R(T_b - T_0)}, \quad u_0 = \frac{T_0}{T_b - T_0}, \quad u_{ig} = \frac{T_{ig} - T_0}{T_b - T_0}. \quad (9)$$

When the radius of cylindrical domain Ω tends to 0, (7) may be reduced to the one-dimensional problem:

$$\frac{\partial u}{\partial t} = \frac{\partial^2 u}{\partial z^2} + (1 - \eta)^N \Phi(u), \quad \frac{\partial \eta}{\partial t} = (1 - \eta)^N \Phi(u). \quad (10)$$

A solution $\{u(t, z), \eta(t, z)\}$ of (10) is called a travelling wave when it is of the form $u(t, z) = \tilde{u}(z - ct)$ and $\eta(t, z) = \tilde{\eta}(z - ct)$ with constant c . A travelling wave corresponds to a combustion wave of steady-state mode, that is, it keeps the profile $\{\tilde{u}(\cdot), \tilde{\eta}(\cdot)\}$ and propagates at the constant velocity c .

Review of mathematical results

We here briefly review mathematical results related to the gasless combustion problem (7). The existence of travelling wave of (10) has been shown by Logak and Loubeau [6] without stability analysis, and Bonnet and Logak [2] has implied the instability of travelling wave for high effective activation energy e in the one-dimensional problem (10).

There are many numerical studies on the one-dimensional problem, where e is adopted as a parameter. The following transitions of propagating patterns have been reported as e is increased: appearance of pulsating waves through the Hopf bifurcation of travelling waves, chaotic behavior after period doubling bifurcations of pulsating waves ([1]), intermittent propagation phenomena ([4]) and so on.

Ivleva et al. [5] have proposed a two-dimensional system of equations on the cylindrical surface, and they have succeeded in numerical computation of two-dimensional helical waves based on their system. On the other hand, Matkowsky and Sivashinsky [7] have shown the destabilization of travelling wave via Hopf bifurcation by using their reduced equation obtained from (5) in the limit where the effective activation energy E tends to the infinity. By the use of the reduced equation, Sivashinsky [11] has implied also the appearance of helical wave through a Hopf-type bifurcation of planar propagating wave.

As stated in the above, the existence of helical wave has been shown only for the two-dimensional problem and the reduced equation. Different from [5] nor [11], we deal with the three-dimensional full system of equations (7) in the present paper. Through our approach, we will obtain the following new insights:

- (1) propagation patterns in the interior of cylindrical domain,
- (2) difference in propagation patterns between the two- and three-dimensional problems,
- (3) dependence of propagation patterns on the effective activation energy E .

Numerical simulation

The reaction order N should equal $m + n$ doubtless if the chemical reaction formula (1) exactly describes the SHS process, however, (1) is a highly idealized and simplified one. In the actual synthesis of TiC for instance, there can occur the dissolution of carbon into a titanium melt and the diffusion of carbon through a solid TiC layer. In order to introduce effects ignored in (1) into our mathematical model, we leave N as a free parameter independent of m and n .

In our numerical simulation of (7) - (8) we decide values of parameters according to experimental data of the TiC combustion synthesis ([3] and [12]):

$$\begin{cases} m = 1, & n = 1, & N = 3.0, & T_0 = 298 \text{ K}, & T_{ig} = 1924 \text{ K}, \\ C_p = 53.78 \text{ J/mol}\cdot\text{K}, & Q_M = Q_{c_0}/\rho = 184.5 \times 10^3 \text{ J/mol}. \end{cases} \quad (11)$$

The above values of C_p and Q_M are the averaged one from 298 K to 3200 K. The value of effective activation energy E is of order 10^5 J/mol, and it depends on the particle sizes of reactants. For this reason we leave E as a controllable parameter in our numerical simulation.

One-dimensional patterns

In numerical simulations of the one-dimensional problem (10), as reported in previous works, we also have observed stable travelling waves for small values of effective activation energy E , stable pulsating waves arising via the Hopf bifurcation of travelling waves for

medium E , pulsating waves after period doubling bifurcation for large E and chaotic combustion waves for much larger E . We note that as E increases the reaction stagnates and the average velocity of propagation becomes slower.

Three-dimensional patterns

We consider (7) in the cylindrical domain $\Omega = \{(r, \theta, z); 0 \leq r < R_0, 0 \leq \theta < 2\pi, 0 < z < L_z\}$ subject to the boundary condition:

$$\frac{\partial u}{\partial z} = 0 \quad \text{for } z = 0, z = L_z \text{ and } t > 0, \quad \frac{\partial u}{\partial r} = 0 \quad \text{for } r = R_0 \text{ and } t > 0. \quad (12)$$

When the value of effective activation energy E is set so that the one-dimensional problem (10) has a stable travelling wave, the planar travelling wave is also stable in the cylindrical domain with various radiuses R_0 in all of our numerical simulations.

We turn to the case where E is selected so that (10) has a stable pulsating wave. The planar pulsating wave is still stable in the cylindrical domain if R_0 is small, however, it loses its stability when R_0 is large enough and helical combustions become dominant for large R_0 . Every helical wave has one or more reaction spots where the temperature is very high. In some cases reaction spots seem to move independently and there occur annihilation of reaction spots by collision and creation of reaction spots (irregular helical waves). In other cases several reaction spots seem to rotate on the cylindrical surface in the same direction (regular helical wave). For the same physical and chemical parameters, there coexist irregular patterns and regular patterns with various numbers of reaction spots. These differences in patterns are caused only by differences in the initial distribution of temperature. The rotation direction also depends on the initial distribution. Among various helical patterns, an irregular one may be most popular in the sense that one has to carefully set the initial distribution in order to get a regular helical pattern while one needs no special care for obtaining an irregular one.

In the isothermal surface of irregular wave, there appear elevation and subsidence erratically as time marches on. An isothermal surface of regular helical wave has some arms of which number is the same as that of reaction spots. The isothermal surface with arms helically rotates down in the cylindrical domain as time passes on. Observing the distribution of temperature on horizontal cross section moving with the corresponding helical wave, we know that a pulsating motion is dominant near the center while a helical rotation is dominant near the cylindrical surface.

Vestiges of helical combustion

The system (7) subject to the boundary condition (12) has helical waves as stated in the above. However, because (12) means that there is no heat loss through the synthesis process, $u(t, \mathbf{x})$ and $\eta(t, \mathbf{x})$ tend to 1 as $t \rightarrow \infty$ for any $\mathbf{x} \in \Omega$ under suitable initial conditions and there is no vestige of helical combustion left at the completion of synthesis. In the actual synthesis, it is hard to keep out heat loss from the cylindrical surface caused by convection and radiation. With a view to checking an effect of heat loss we replace the

second condition in (12) with the following simple heat loss condition by convection

$$\frac{\partial u}{\partial r} = -\lambda u \quad \text{for } r = R_0 \text{ and } t > 0, \quad (13)$$

where λ is a positive constant. Just like the case of no heat loss boundary condition, the temperature distribution becomes spatially uniform as $t \rightarrow \infty$. Nevertheless, vestiges of helical combustion are remained in the distribution of synthesis rate η . We thus succeed in the reproduction of vestiges of helical combustion similar to those observed in laboratory.

Differences between two- and three-dimensional patterns

We have carried out numerical simulations of the following plain two-dimensional problem

$$\begin{aligned} \frac{\partial u}{\partial t} = \frac{\partial^2 u}{\partial \theta^2} + \frac{\partial^2 u}{\partial z^2} + (1 - \eta)^N \Phi(u), \quad \frac{\partial \eta}{\partial t} = (1 - \eta)^N \Phi(u) \\ \text{for } \theta \in (0, L_\theta), z \in (0, L_z) \text{ and } t > 0 \end{aligned} \quad (14)$$

subject to the periodic boundary condition in the θ -direction and the no-flux boundary condition at $z = 0$ and $z = L_z$. The system (14) has helical waves in the same way as the three-dimensional problem, that is, helical waves become the most popular type of solutions of (14) when E is set so that (10) has a pulsating wave and L_θ is large enough. Different from the three-dimensional case, the two-dimensional system prefers regular helical waves in the sense that solutions of (14) tend to a regular helical wave as time goes on for a very large family of initial conditions.

Discussion

One of our purpose in the present paper is to make clear the organizing center of helically propagating waves. We propose a conjecture that the organizing center is the existence of stable pulsating wave of the corresponding one-dimensional problem, although a mathematically rigorous proof has not been obtained. This conjecture is true for the model (7) as described in the above. With the view of confirming the conjecture, we consider an autocatalytic reaction model. The one-dimensional model is written as

$$\frac{\partial u}{\partial t} = \frac{\partial^2 u}{\partial z^2} + v f(u), \quad \frac{\partial v}{\partial t} = d \frac{\partial^2 v}{\partial z^2} - v f(u), \quad (15)$$

where $f(u) = u^m$ ($m \geq 1$) and $d \geq 0$ is a constant, and Metcalf et al. [8] have proved that (15) has travelling waves in the case of $d = 0$. Metcalf et al. have also studied numerically the behavior of solutions of (15) for various m and d , and it has been reported that (15) has a stable travelling wave for some m and d while a travelling wave loses its stability and a pulsating wave emerges for some other m and d . We have carried out numerical simulation of the two-dimensional autocatalytic reaction model:

$$\begin{aligned} \frac{\partial u}{\partial t} = \frac{\partial^2 u}{\partial \theta^2} + \frac{\partial^2 u}{\partial z^2} + v f(u), \quad \frac{\partial v}{\partial t} = d \left(\frac{\partial^2 v}{\partial \theta^2} + \frac{\partial^2 v}{\partial z^2} \right) - v f(u) \\ \text{for } \theta \in (0, L_\theta), z \in (0, L_z) \text{ and } t > 0 \end{aligned} \quad (16)$$

subject to the periodic boundary condition in the θ -direction and the no-flux boundary condition at $z = 0$ and $z = L_z$, and have found the following properties supporting the above mentioned conjecture:

- (a) When (15) has a stable pulsating wave, the planar pulsating wave is still stable in the two-dimensional domain $(0, L_\theta) \times (0, L_z)$ if L_θ is small while a helical wave becomes dominant for large L_θ .
- (b) When (15) has a stable travelling wave, the planar travelling wave is also stable in the two-dimensional domain with all the values of L_θ we tried.

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